

# Reduced equations of motion for quantum systems driven by diffusive Markov processes

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The expansion of a stochastic Liouville equation for the coupled evolution of a quantum system and an Ornstein-Uhlenbeck process into a hierarchy of coupled differential equations is a useful technique that simplifies the simulation of stochastically-driven quantum systems. We expand the applicability of this technique by completely characterizing the class of diffusive Markov processes for which a useful hierarchy of equations can be derived. The expansion of this technique enables the examination of quantum systems driven by non-Gaussian stochastic processes with bounded range. We present an application of this extended technique by simulating Stark-tuned Förster resonance transfer in Rydberg atoms with non-perturbative position fluctuations.

Describing the dynamics of a quantum system coupled to uncontrolled degrees of freedom has been an important problem since the inception of quantum mechanics, e.g., [1]. The accurate description of such *open quantum systems* is particularly vital for the design of quantum technologies, such as quantum computers. Several approximate and exact methods exist for describing the dynamics of open quantum systems, including master equations, surrogate Hamiltonians, and Monte-Carlo numerical simulations.

In this work, we examine quantum systems driven linearly by classical fluctuations. That is, the Hamiltonian for the system is described by  $\hat{H}_\Omega(t) = \hat{H}_0 + \Omega(t)\hat{V}$ , where  $\Omega(t)$  is a time-dependent stochastic variable. This is a sub-class of a more general open quantum system where  $\Omega(t)$  is an operator in the Hilbert space of an uncontrolled environment. The replacement of the operator with a scalar variable is an approximation that is valid in certain limits (e.g., in the high temperature limit of a bosonic environment). Scalar fluctuations can also describe noise in a quantum system that is controlled by an effectively classical quantity, such as a gate voltage.

For such stochastic evolution, the dynamics of the system under a *given* trace of the noise is dictated by the von-Neumann (Schrödinger) equation:

$$\frac{\partial}{\partial t} \hat{\rho}(t|\{\Omega(t)\}) = -\frac{i}{\hbar} [\hat{H}_\Omega(t), \hat{\rho}(t|\{\Omega(t)\})]. \quad (1)$$

Here, the notation  $\hat{\rho}(t|\{\Omega(t)\})$  is used to explicitly indicate that this density matrix is *conditioned* on a particular realization of the noise process. This equation is formally solved to yield  $\hat{\rho}(t|\{\Omega(t)\}) = \hat{U}(t, 0) \hat{\rho}^0 \hat{U}(t, 0)^\dagger$ , for initial state  $\rho^0$ , where  $\hat{U}(t, 0) = \mathcal{T} e^{-\frac{i}{\hbar} \int_0^t \hat{H}_\Omega(t) dt}$  with  $\mathcal{T}$  being the time-ordering operator. We are often more interested in the unconditioned evolution of the system state, after the fluctuating quantity has been averaged:

$$\hat{\rho}(t) = \langle \hat{\rho}(t|\{\Omega(t)\}) \rangle_{\{\Omega(t)\}}, \quad (2)$$

where the angled brackets denote an expectation value over the stochastic process up to time  $t$ . We will refer to differential equations describing the evolution of

this averaged quantity as *reduced equations of motion*. In this work we derive reduced equations of motion for a quantum system coupled to a wide family of stochastic processes.

We focus on stochastic processes that are time-continuous, time-homogenous, and Markov, which means that the evolution of the conditional probability distribution for the process evolves as [2–4]

$$\frac{\partial}{\partial t} P(\Omega, t|\Omega', t') = \Gamma_\Omega P(\Omega, t|\Omega', t'), \quad (3)$$

where  $P(\Omega, t|\Omega', t')$  is the probability that the stochastic process takes the value  $\Omega$  at time  $t$  given that it took the value  $\Omega'$  at time  $t'$  ( $t' \leq t$ ).  $\Gamma_\Omega$  is the forward *generator* of the process and is a differential operator only involving derivatives with respect to  $\Omega$ . The generator of evolution is in general a complex quantity that results from a *Kramers-Moyal* expansion of the (classical) master equation for the probability distribution of the process [2]. Below, we will restrict ourselves to certain forms of this generator, but for now only the Markov assumption will be used.

In order to derive an expression related to the reduced equations of motion in a simple manner, consider the quantity  $\hat{\rho}(t, \Omega) \equiv \hat{\rho}_\Omega(t) P(\Omega, t)$ , which is the joint distribution of the quantum-mechanical coordinates and the stochastic variable at time  $t$ .  $\hat{\rho}_\Omega(t) \equiv \hat{\rho}(t|\Omega(t))$  is the density matrix conditioned on the value of the stochastic process at time  $t$  (in contrast to  $\hat{\rho}(t|\{\Omega(t)\})$ , which is the density matrix conditioned on an entire history of the stochastic process). Evaluating the time derivative of this joint distribution results in the *stochastic Liouville equation*, first derived by Kubo [5]:

$$\frac{\partial \hat{\rho}(t, \Omega)}{\partial t} = \left[ -\frac{i}{\hbar} \hat{H}_0^\times - \frac{i}{\hbar} \Omega(t) \hat{V}^\times \right] \hat{\rho}(t, \Omega) + \Gamma_\Omega \hat{\rho}(t, \Omega) \quad (4)$$

where  $A^\times B \equiv [A, B]$ . Here, we have used the fact that because the stochastic process is Markov, the time evolution of  $P(\Omega, t)$  follows the same law as the conditional distribution in Eq. (3) and is generated by  $\Gamma_\Omega$ . This simple linear form for the evolution of the joint distribution is possible because both the conditional quantum

density matrix and the probability distribution for the stochastic variable evolve linearly and in a Markov fashion. Because  $\hat{\rho}(t, \Omega)$  is a joint distribution, its marginal over the stochastic variable yields the average density matrix, i.e.,  $\hat{\rho}(t) = \int_{\mathcal{D}} d\Omega \hat{\rho}(t, \Omega)$ , where  $\mathcal{D}$  is the range of the stochastic variable [6].

**Markov diffusion processes.** A wide a class of physical processes can be approximated by truncating the Kramers-Moyal expansion for the generator  $\Gamma_{\Omega}$  of the Markov process at the second term [2]. This results in the *Fokker-Planck* equation for the probability distribution, with the generator being the differential operator [4]:  $\Gamma_{\Omega}^{\text{FP}} \equiv \Phi_{\Omega} = -\frac{\partial}{\partial \Omega} A(\Omega) + \frac{1}{2} \frac{\partial^2}{\partial \Omega^2} B(\Omega)$ , where  $A(\Omega)$  and  $B(\Omega)$  are real differentiable coefficients, with the restriction that  $B > 0$ .

For such *diffusion processes*, it is common to define the backward, or adjoint, generator for the process:  $\overleftarrow{\Phi}_{\Omega} = A(\Omega) \frac{\partial}{\partial \Omega} + \frac{1}{2} B(\Omega) \frac{\partial^2}{\partial \Omega^2}$ . For a Markov process, Eq. (3) is often called the *forward Kolmogorov equation* and  $\frac{\partial}{\partial t'} P(\Omega, t | \Omega', t') = \overleftarrow{\Phi}_{\Omega'} P(\Omega, t | \Omega', t')$  is the *backward Kolmogorov equation*. Let  $f_n(\Omega)$  and  $\overleftarrow{f}_n(\Omega)$  be eigenfunctions of the forward and backward generators, respectively, i.e.,  $\Phi_{\Omega} f_n(\Omega) = -\lambda_n f_n(\Omega)$  and  $\overleftarrow{\Phi}_{\Omega} \overleftarrow{f}_n(\Omega) = -\lambda_n \overleftarrow{f}_n(\Omega)$ , with  $\lambda_n \geq 0$  [2]. These two eigenfunctions are related through the stationary distribution of the process, explicitly  $f_n(\Omega) = P^0(\Omega) \overleftarrow{f}_n(\Omega)$ , where  $P^0$  is the stationary distribution of the process  $\Omega$  [3]. Furthermore, the eigenfunctions of the backward generator form a complete, orthogonal system (on the range  $\mathcal{D}$ ) under the measure induced by the stationary distribution of the process:  $\int_{\mathcal{D}} d\Omega P^0(\Omega) \overleftarrow{f}_n(\Omega) \overleftarrow{f}_m(\Omega) = \delta_{mn}$ , which in turn implies the following about the eigenfunctions of the forward generator [3]:  $\int_{\mathcal{D}} d\Omega (P^0(\Omega))^{-1} f_n(\Omega) f_m(\Omega) = \delta_{mn}$

**Diffusive hierarchical equations of motion.** Us-

ing the completeness and orthogonality of the backward and forward generator eigenfunctions [3], we can expand  $P(\Omega, t)$  in terms of these eigenfunctions, and consequently,  $\hat{\rho}(t, \Omega) \equiv \hat{\rho}_{\Omega}(t) P(\Omega, t) = \sum_{n=0}^{\infty} \bar{\rho}_n(t) f_n(\Omega)$ . Here,  $\bar{\rho}_n(t)$  are unnormalized auxiliary density matrices, which do not have the interpretation of being *conditioned* density matrices of the quantum system. Then, expressing the stochastic Liouville equation Eq. (4) in terms of this eigenfunction expansion yields

$$\sum_{n=0}^{\infty} \frac{\partial}{\partial t} \bar{\rho}_n(t) f_n(\Omega) = \sum_{n=0}^{\infty} -\frac{i}{\hbar} H_0^{\times} \bar{\rho}_n(t) f_n(\Omega) - \frac{i}{\hbar} V^{\times} \bar{\rho}_n(t) [\Omega(t) f_n(\Omega)] - \lambda_n \bar{\rho}_n(t) f_n(\Omega). \quad (5)$$

To simplify further, we make the assumption that the eigenfunctions of the backward Markov generator are polynomials in  $\Omega$ , i.e.,  $\overleftarrow{f}_n(\Omega)$  is an  $n^{\text{th}}$  order polynomial in  $\Omega$ . With this assumption, we utilize the following theorem, typically attributed to Favard [7], which states an important property of orthogonal polynomials:

**Theorem (Favard):** Let  $\{p_n(x)\}, n \geq 0$  be a system of polynomials. This system satisfies a three-term recurrence relation  $p_{n+1}(x) = (A_n x + B_n) p_n(x) - C_n p_{n-1}(x)$ , (with  $p_{-1}(x) = 0$ ) if and only if it is a system of *orthogonal polynomials*. Here  $A_n \neq 0, B_n$  and  $C_n \neq 0$  are  $\Omega$ -independent (real) recurrence coefficients.

Favard's theorem implies that the orthogonal polynomial eigenfunctions of the backward Markov generator (and hence, those of the forward Markov generator) satisfy the three-term recurrence relations:

$$\Omega f_n(\Omega) = \frac{C_n}{A_n} f_{n-1}(\Omega) - \frac{B_n}{A_n} f_n(\Omega) + \frac{1}{A_n} f_{n+1}(\Omega). \quad (6)$$

Using this recurrence relation, the eigenfunction expansion of the stochastic Liouville equation becomes:

$$\sum_{n=0}^{\infty} \frac{\partial}{\partial t} \bar{\rho}_n(t) f_n(\Omega) = \sum_{n=0}^{\infty} \left[ -\frac{i}{\hbar} H_0^{\times} - \lambda_n \right] \bar{\rho}_n(t) f_n(\Omega) - \frac{i}{\hbar} V^{\times} \bar{\rho}_n(t) \left[ \frac{1}{A_n} f_{n+1}(\Omega) + \frac{C_n}{A_n} f_{n-1}(\Omega) - \frac{B_n}{A_n} f_n(\Omega) \right] \quad (7)$$

Multiplying both sides of this equation by  $f_m(\Omega)/P^0(\Omega)$  and integrating over  $\Omega$  results in an equation for each  $m$  because of the orthogonality of the functions  $f_n(\Omega)$ . These equations form a hierarchy of coupled (operator) differential equations, which we refer to as *diffusive hierarchical equations of motion* (DHEOM) [8]:

$$\begin{aligned} \frac{\partial}{\partial t} \bar{\rho}_0(t) &= - \left[ \frac{i}{\hbar} \left( H_0^{\times} - \frac{B_0}{A_0} V^{\times} \right) + \lambda_0 \right] \bar{\rho}_0(t) - \frac{i}{\hbar} \frac{C_1}{A_1} V^{\times} \bar{\rho}_1(t), \\ \frac{\partial}{\partial t} \bar{\rho}_n(t) &= - \left[ \frac{i}{\hbar} \left( H_0^{\times} - \frac{B_n}{A_n} V^{\times} \right) + \lambda_n \right] \bar{\rho}_n(t) - \frac{i}{\hbar} \frac{C_{n+1}}{A_{n+1}} V^{\times} \bar{\rho}_{n+1}(t) - \frac{i}{\hbar} \frac{1}{A_{n-1}} V^{\times} \bar{\rho}_{n-1}(t), \quad n > 0. \end{aligned} \quad (8)$$

These equations are useful because they describe the evolution of the system density matrix under the influence of stochastic noise, but without explicit reference to noise variables. In addition, each member of the hierarchy only

couples to its two neighbors, i.e.,  $n$  couples to  $n+1$  and  $n-1$ , making their integration easy. We also need to specify the initial conditions and the prescription for calculating the system density matrix from the solution of the

hierarchy of coupled equations. A physically motivated initial state for the quantity  $\hat{\rho}(t, \Omega)$  is  $\hat{\rho}(0, \Omega) = \hat{\rho}^0 P^0(\Omega)$ , where  $P^0$  is the stationary (equilibrium) distribution of the noise process and  $\hat{\rho}^0$  is any system density matrix. Evolution from this initial state describes the response of a quantum system to fluctuations around the bath equilibrium. Now,  $\Phi_\Omega P^0 = 0$  by definition, and therefore,  $\bar{\rho}_0(0) = \hat{\rho}^0$ , and  $\bar{\rho}_n(0) = 0 \forall n \neq 0$ . At any time, the system density matrix is defined as the integral of the quantity  $\hat{\rho}(t, \Omega)$  over  $\Omega$ :

$$\hat{\rho}(t) = \int_{\mathcal{D}} d\Omega \hat{\rho}(t, \Omega) = \int_{\mathcal{D}} d\Omega \sum_{n=0}^{\infty} \bar{\rho}_n(t) f_n(\Omega) \propto \bar{\rho}_0(t).$$

The proportionality is a result of the following property of the eigenfunctions:

$$\int_{\mathcal{D}} d\Omega f_n(\Omega) \propto \int_{\mathcal{D}} d\Omega P^0(\Omega) \overleftarrow{f}_n(\Omega) \overleftarrow{f}_0(\Omega) = \delta_{n0},$$

where this proportionality follows from the fact that  $\overleftarrow{f}_0$  is a zeroth-order polynomial in  $\Omega$ . As we shall see below, it is always possible to choose  $\overleftarrow{f}_0 = 1$ , and therefore, the

proportionality above can be converted to an equality. Hence, the auxiliary density matrix  $\bar{\rho}_0(t)$  always keeps track of the averaged system density matrix  $\hat{\rho}(t)$ .

To summarize, we have shown that the dynamics of a quantum system that is linearly driven by a diffusion process with a Markov generator possessing polynomial eigenfunctions can be described by a hierarchy of coupled dynamical equations. The solution to these equations will reproduce the reduced density matrix of the quantum system at any time, with no approximations. However, the above hierarchy of equations is infinite, and therefore, we require some truncation strategy in order to solve them. If the equations can be truncated at some  $n = N$  at the expense of bounded error, they can be numerically solved (or analytically solved via a partial fraction expansion if  $H_0$  is time-independent [9]). A general truncation strategy exists if the following conditions hold: (1) for large enough  $n = N$ ,  $|\lambda_N| \gg \|H_0^\times - \frac{B_N}{A_N} V^\times\|_2$ , and (2)  $\frac{1}{A_n} \in \omega\left(\frac{C_n}{A_n}\right)$ , where  $\|\cdot\|_2$  is the induced 2-norm and  $f_n \in \omega(g_n)$  indicates that  $f_n$  dominates  $g_n$  asymptotically (in  $n$ ). In appendix B, we develop such a general truncation strategy that results in the following *terminator equation* for the hierarchy at a level  $N$  satisfying these conditions:

$$\frac{\partial}{\partial t} \bar{\rho}_N(t) = - \left[ \frac{i}{\hbar} \left( H_0^\times - \frac{B_N}{A_N} V^\times \right) + \lambda_N \right] \bar{\rho}_N(t) - \frac{1}{\hbar^2} \frac{C_{N+1}}{\lambda_{N+1} A_{N+1} A_N} V^\times V^\times \bar{\rho}_N(t) - \frac{i}{\hbar} \frac{1}{A_{N-1}} V^\times \bar{\rho}_{N-1}(t). \quad (9)$$

We now turn to characterizing the diffusive stochastic processes with generators that have polynomial eigenfunctions. A quantum system driven by each one of these processes will have a dynamical description in terms of the DHEOM derived above. The combination of Bochner's theorem [10], which is stated explicitly in appendix A, and Favard's theorem implies that there are only three diffusive processes with backward and forward generators that have orthogonal polynomial eigenfunctions: (a) the Ornstein-Uhlenbeck (OU) process, defined on  $(-\infty, \infty)$  (b) the square-root process, defined on  $(\Omega_{\min}, \infty)$  and (c) the Jacobi process, defined on  $(\Omega_{\min}, \Omega_{\max})$ . In table I of appendix A, we list these diffusive processes with their orthogonal polynomial eigenfunctions and other relevant properties. In appendix C, we also explicitly write the DHEOM that describe the dynamics of a quantum system linearly driven by each of the above three stochastic processes. The Jacobi process is particularly efficient to simulate because the eigenvalues  $\lambda_n$  of its generator scale quadratically with  $n$ , making truncation possible at a smaller depth than for the other two processes, where  $\lambda_n$  scales linearly with  $n$ . We note that the DHEOM for the OU process was previously derived by Kubo and Tanimura [5, 9], but this more general framework resulting in DHEOMs for all major diffusive Markov processes was heretofore unknown. For reasons

explained in appendix C, the DHEOM method is limited to simulating square-root processes with mean reversion rate  $\gamma > 1$ , whereas any  $\gamma$  is valid for the other processes. Finally, for completeness appendix D presents the DHEOM for the propagator for the dynamical map as opposed to the density matrix.

**Application and Discussion.** The primary advantage of our general formulation of DHEOM is that it allows for the efficient simulation of quantum systems driven by noise sources with bounded range and non-Gaussian amplitude distribution. As an application, we consider energy transfer between two systems mediated by a dipole-dipole interaction. This interaction is an approximation to the Coulomb coupling of electric charge distributions and is the basis of several common experimental techniques, such as Förster resonance energy transfer (FRET) spectroscopy [11] and Stark-tuned resonant transfer in Rydberg atoms [12, 13]; we focus on this latter system as our example.

The electric dipole-dipole coupling energy scales as  $J \propto 1/R^3$ , where  $R$  is the distance between the coupled molecules or atoms. Consider the case where this interparticle spacing is varying in time as a result of thermal, electromagnetic, or vibrational fluctuations. If the fluctuations in  $R$  are treated perturbatively the dipole-dipole coupling energy can be expanded linearly around

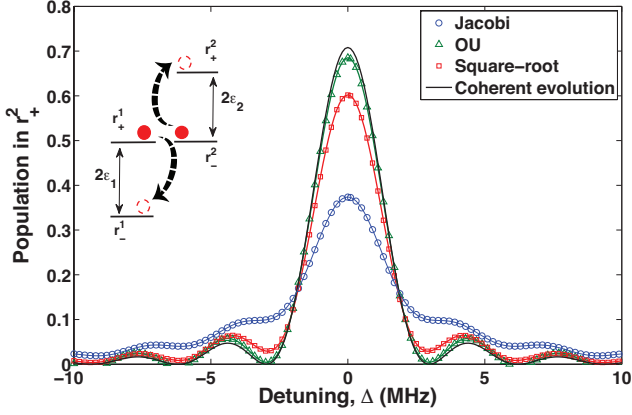


FIG. 1. Average population in the excited state of atom 2 after an interaction time of  $T = 1\mu\text{s}$  as a function of the Stark detuning for different models of motional noise. The black (solid) curve is for coherent evolution with no fluctuation in inter-atom distance. The parameters used are:  $\gamma = 1.5$ ,  $\mu = 1$  for all noise sources, and  $\sigma^2 = 0.3$  for the OU process. The square root process is defined in the semi-interval  $[0, \infty)$  with  $c_1 = 1$ , and the Jacobi process in the interval  $(1/8, 8)$  with  $c = 1$ . See appendix A for the interpretation of these parameters.

the mean separation,  $R_0$ :  $J \sim 1/R_0^3 - (3/R_0^3)\delta R(t)$ . However, in cases where such a perturbative treatment is not accurate we must consider the energy  $J$  as a time-dependent fluctuating quantity in a range  $[J_{\min}, J_{\max}]$ . To explore the consequences of such a non-perturbative treatment, consider the Hamiltonian of two interacting Rydberg atoms:  $H = -\epsilon_1\sigma_z^1 - \epsilon_2\sigma_z^2 + J(t)(\sigma_+^1\sigma_-^2 + \sigma_-^1\sigma_+^2)$ , where we restrict the description of the atoms to the two energy levels relevant to the energy transfer, and therefore use Pauli matrices to describe them as effective two-level systems in the basis  $|r_-^i\rangle, |r_+^i\rangle$  where  $r_\pm^i$  are Rydberg states for atom  $i$  with the energy gap  $\epsilon_i$ , i.e.,  $\sigma_z^i|r_\pm^i\rangle = \pm\epsilon_i|r_\pm^i\rangle$ . Typically, these levels are chosen so that when an electric field tunes atomic energies by the Stark effect they satisfy  $\epsilon_1 = \epsilon_2$  (Förster resonance condition) at some critical field value. For example, in Ref. [13],  $|r_+^1\rangle = |37P_{3/2}\rangle, |r_-^1\rangle = |37S_{1/2}\rangle, |r_+^2\rangle = |38S_{1/2}\rangle, |r_-^2\rangle = |37P_{3/2}\rangle$ , where these are fine states of Rb atoms. We ignore relaxation of these states because for sufficiently cooled Rydberg atoms it can be neglected on the timescales we are considering [14]. We also assume that the relative orientation of the atomic dipoles remains constant. The initial state is the excited state of atom 1:  $\rho^0 = |r_+^1, r_-^2\rangle\langle r_+^1, r_-^2|$ , and the dipole-dipole coupling is assumed to fluctuate as  $J(t) \equiv J_0\Omega(t)$ , where  $\Omega(t)$  is one of the three diffusive noise processes described above. The quantity being observed (e.g., by selective field ionization [13]) is the population in the excited state of atom 2 after an interaction time  $T$ . Although position fluctuations were reported to be minor in the experiment of Ref. [13], we use physical parameters compatible with this experiment for concreteness:  $J_0/\hbar = 0.5\text{MHz}$ , and

$T = 1\mu\text{s}$ . Fig. 1 shows the average population of  $|r_+^2\rangle$  at  $T$  for different values of the Stark detuning  $\Delta \equiv \epsilon_2 - \epsilon_1$ . We expect that this transferred population will drop off as  $|\Delta|$  increases (as the atoms become less resonant), but the figure shows that the behavior of this drop-off depends heavily on the exact nature of the fluctuations in  $R$ , although all processes have the same mean  $\mu = 1$  and mean reversion rate  $\gamma = 1.5$ . The OU and square-root processes predict average populations that have significant oscillations as a function of the detuning, similar to the completely coherent (noiseless) case. In contrast, the Jacobi process driven evolution predicts damped oscillations and smaller transfer populations. We predict that for experiments performed in this regime, where position fluctuations are relevant, the Jacobi process average will be more accurate (and they are certainly more consistent with what is experimentally observed in Ref. [13]). This is because the Jacobi process is a more accurate description than the OU and square-root processes, which can only be approximations in this scenario; realistic trapping conditions impose strict lower and upper bounds on  $J$  resulting from the fact that the atoms can only drift within the trap volumes. The OU and square-root simulations predict large transfer populations because rare, unphysical, large magnitude couplings contribute to the averages in these cases while they do not to the Jacobi process average. These results emphasize the advantage of being able to efficiently average over non-Gaussian noise processes with bounded range. To underscore the numerical efficiency of the DHEOM simulations, we note that the calculations producing Fig. 1 were performed 10 times faster using the present theory than conventional Monte Carlo sampling of the noise processes [15]. Furthermore, because of the well-defined truncation strategy, convergence issues and noise are not a concern for DHEOM simulations as in the Monte Carlo approach.

**Summary.** We have completely characterized the class of diffusively driven quantum systems for which a useful hierarchy of reduced equations of motion can be derived, and explicitly derived these DHEOM. We applied the technique to examine position fluctuation dependence of Stark tuned Förster resonance between Rydberg atoms. We expect that the general techniques developed in this work will be useful for widening the range of open quantum systems that can be simulated efficiently. Use of the DHEOM for Jacobi processes will be particularly useful for modeling quantum systems subject to fluctuations with bounded range, e.g., classical noise in external control fields [16].

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## Appendix A: Bochner's theorem and polynomial eigenfunctions

In the main text, we claim that there are three stochastic processes that admit an eigenfunction of their Markov generator that is an orthogonal polynomial. This result follows from Bochner's theorem [10]:

*Theorem (Bochner):* Consider the eigenvalue problem

$$f(x)\frac{d^2}{dx^2}y(x) + g(x)\frac{d}{dx}y(x) + h(x)y(x) = \lambda y(x) \quad (\text{A1})$$

where  $f, g, h$  are fixed polynomials and  $\lambda$  is independent of  $x$ . There are only five classes of polynomial solutions to this problem, where  $y_n(x)$  is a polynomial of degree  $n$ , and they are:

1.  $y_n(x) = \text{He}_n(x)$ ,  $n \in \mathbb{N}_0$  is a Hermite polynomial, defined on the infinite interval  $x \in (-\infty, \infty)$ .
2.  $y_n(x) = L_n^{(\alpha)}(x)$ ,  $n \in \mathbb{N}_0$  is a generalized Laguerre polynomial, defined on the semi-infinite interval  $x \in [a, \infty)$  or  $x \in (-\infty, a]$  for  $a \in \mathbb{R}$ .
3.  $y_n(x) = J_n^{(\alpha, \beta)}(x)$ ,  $n \in \mathbb{N}_0$  is a Jacobi polynomial, defined on the interval  $x \in (a, b)$  for  $a, b \in \mathbb{R}$ .
4.  $y_n(x)$  is in the set  $\{x^n\}$  for  $n \in \mathbb{N}_0$ .
5.  $y_n(x) = B_n(x)$  is a Bessel polynomial, defined on the infinite interval  $x \in (-\infty, \infty)$ .

Note that the backward Kolmogorov equation is an example of the differential equation considered in Eq. (A1), with the additional restriction that  $f(x) > 0$ . Therefore, all polynomial eigenfunctions of the backwards Kolmogorov generator fall within the Bochner classes identified above. However, since we also require that our eigenfunctions satisfy a three-term recurrence relation Favard's theorem further restricts us to the subset of the Bochner classes that are orthogonal polynomials. The Hermite, Laguerre, and Jacobi polynomials are all classical orthogonal polynomials and therefore obviously orthogonal. The polynomials sequence in class 4 above is not an orthogonal sequence, and similarly, the Bessel polynomials are not an orthogonal set of polynomials for any positive-definite measure. The Bessel polynomials are orthogonal under a quasi-definite measure and hence do satisfy a three-term recurrence relation [10]. However the positive-definiteness of the measure is an important feature in the following and therefore we will not consider the Bessel polynomials. Therefore, the combination Favard's and Bochner's theorems imply that there are only three diffusive processes with backward and forward generators that have orthogonal polynomial eigenfunctions: (a) the Ornstein-Uhlenbeck process, (b) the square-root (or Cox-Ingersoll-Ross) process, and (c) the Jacobi process. This result has been derived by a number of authors in the past, in different mathematical contexts, including in Refs. [19–21].

## Appendix B: Truncation of DHEOM

The conditions for a general truncation strategy to exist for the DHEOM are: (1) for large enough  $n = N$ ,  $|\lambda_N| \gg \|H_0^\times - \frac{B_N}{A_N}V^\times\|_2$ , and (2)  $\frac{1}{A_n} \in \omega\left(\frac{C_n}{A_n}\right)$ , where  $\|\cdot\|_2$  is the operator 2-norm and  $f_n \in \omega(g_n)$  indicates that  $f_n$  dominates  $g_n$  asymptotically (in  $n$ ). To develop the truncation strategy under these conditions we follow Ref. [9] and consider the formal solution to the  $n = N + 1 \gg 1$  member of the hierarchy:

$$\bar{\rho}_{N+1}(t) = \frac{-i}{\hbar} \int_0^t d\tau \exp\left\{-\left[\frac{i}{\hbar}\left(H_0^\times - \frac{B_{N+1}}{A_{N+1}}V^\times\right) + \lambda_{N+1}\right](t - \tau)\right\} V^\times \left[\frac{C_{N+2}}{A_{N+2}}\bar{\rho}_{N+2}(\tau) + \frac{1}{A_N}\bar{\rho}_N(\tau)\right] \quad (\text{B1})$$

The first condition above implies that by choice of  $N$  we have  $|\lambda_{N+1}| \gg \|H_0 - \frac{B_{N+1}}{A_{N+1}}V\|$ , and we can replace  $\bar{\rho}_i(\tau)$  with  $\bar{\rho}_i(t)$  in the integrand because the exponential term decays much faster than the rate at which  $\bar{\rho}_i(\tau)$  changes. This approximation enables the integral to be computed and results in the solution

$$\bar{\rho}_{N+1}(t) \approx \frac{-i}{\hbar\lambda_{N+1}} V^\times \left[\frac{C_{N+2}}{A_{N+2}}\bar{\rho}_{N+2}(t) + \frac{1}{A_N}\bar{\rho}_N(t)\right]$$

TABLE I. Summary of diffusion processes with backward generators possessing polynomial eigenfunctions.  $I_{(a,b)}$  is the indicator function for the interval  $(a,b)$ ,  $\Gamma[\cdot]$  is the Gamma function, and  $B[\cdot]$  is the Beta function. All processes have exponentially decaying correlation functions, proportional to  $e^{-\gamma t}$ . Note that in order to get convenient recurrence relations, we choose unconventional normalizations for the orthogonal polynomial eigenfunctions.

<b>Ornstein-Uhlenbeck process</b>	
Range of stochastic process $\mathcal{D}_{OU} : -\infty < \Omega < \infty$	
Generator parameters $A^{OU}(\Omega) = -\gamma(\Omega - \mu), \quad \gamma > 0, \mu \in \mathcal{D}_{OU}$ $B^{OU}(\Omega) = \sigma^2$	
Eigenfunctions of backward generator Scaled Hermite polynomials $\overleftarrow{\phi}_n(\Omega) \equiv \frac{1}{n!} \text{He}_n(\sqrt{\frac{2\gamma}{\sigma^2}}(\Omega - \mu)) = \sum_{m=0}^{\lfloor n/2 \rfloor} \frac{(-1)^m}{m! 2^m (n-2m)!} \left( \sqrt{\frac{2\gamma}{\sigma^2}}(\Omega - \mu) \right)^{n-2m}$	
Eigenvalues of backward generator $\overleftarrow{\Phi}_\Omega^{OU} \overleftarrow{\phi}_n(\Omega) = -n\gamma \overleftarrow{\phi}_n(\Omega); \quad \lambda_n = n\gamma$	
Stationary distribution Gaussian distribution $P_0^{OU}(\Omega) = \frac{1}{\sqrt{\pi\sigma^2/\gamma}} e^{-\frac{\gamma(\Omega-\mu)^2}{\sigma^2}}$	
<b>Square-root (or Cox-Ingersoll-Ross) process</b>	
Range of stochastic process $\mathcal{D}_{SR} : -\frac{c_0}{c_1} < \Omega < \infty, \quad c_1 > 0$	
Generator parameters $A^{SR}(\Omega) = -\gamma(\Omega - \mu), \quad \gamma > 0, \mu \in \mathcal{D}_{SR}$ $B^{SR}(\Omega) = c_1\Omega + c_0$	
Eigenfunctions of backward generator Scaled generalized Laguerre polynomials $\overleftarrow{\chi}_n(\Omega) \equiv \frac{1}{n!} L_n^{(\alpha)} \left[ \frac{2\gamma}{c_1} \left( \Omega + \frac{c_0}{c_1} \right) \right] = \frac{1}{n!} \sum_{m=0}^n \binom{n+\alpha}{n-m} \frac{\left( \frac{-2\gamma}{c_1} \left( \Omega + \frac{c_0}{c_1} \right) \right)^m}{m!}$ where $\alpha \equiv \frac{2\gamma}{c_1} \left( \mu + \frac{c_0}{c_1} \right) - 1$	
Eigenvalues of backward generator $\overleftarrow{\Phi}_\Omega^{SR} \overleftarrow{\chi}_n(\Omega) = -n\gamma \overleftarrow{\chi}_n(\Omega); \quad \lambda_n = n\gamma$	
Stationary distribution Gamma distribution $P_0^{SR}(\Omega) = \frac{\left( \frac{2\gamma}{c_1} \right)^{\alpha+1}}{\Gamma[\alpha+1]} e^{-\frac{2\gamma}{c_1} \left( \Omega + \frac{c_0}{c_1} \right)} \left( \Omega + \frac{c_0}{c_1} \right)^\alpha I_{(-\frac{c_0}{c_1}, \infty)}$	
<b>Jacobi process</b>	
Range of stochastic process $\mathcal{D}_J : \omega_1 < \Omega < \omega_2$	
Generator parameters $A^J(\Omega) = -\gamma(\Omega - \mu), \quad \gamma > 0, \mu \in \mathcal{D}_J$ $B^J(\Omega) = -c(\Omega - \omega_1)(\Omega - \omega_2), \quad c > 0$	
Eigenfunctions of backward generator Scaled Jacobi polynomials $\overleftarrow{\zeta}_n(\Omega) \equiv \frac{1}{n!} J_n^{(\alpha, \beta)} \left( \frac{2(\Omega - \omega_2)}{\omega_2 - \omega_1} + 1 \right) = \frac{1}{n!} \sum_{m=0}^n \binom{n}{m} \frac{\Gamma[\alpha + \beta + n + m + 1]}{\Gamma[\alpha + m + 1]} \left( \frac{\Omega - \omega_2}{\omega_2 - \omega_1} \right)^m$ where $\alpha \equiv \frac{2\gamma}{c} \frac{\omega_2 - \mu}{\omega_2 - \omega_1} - 1$ , and $\beta \equiv \frac{2\gamma}{c} \frac{\mu - \omega_1}{\omega_2 - \omega_1} - 1$	
Eigenvalues of backward generator $\overleftarrow{\Phi}_\Omega^J \overleftarrow{\zeta}_n(\Omega) = (-n\gamma - \frac{1}{2}cn(n-1)) \overleftarrow{\zeta}_n(\Omega); \quad \lambda_n = n\gamma + \frac{1}{2}cn(n-1)$	
Stationary distribution Beta distribution $P_0^J(\Omega) = \frac{1}{B[\beta+1, \alpha+1]} \frac{(\Omega - \omega_1)^\beta (\omega_2 - \Omega)^\alpha}{(\omega_2 - \omega_1)^{2\gamma/c - 1}} I_{(\omega_1, \omega_2)}$	

Now the second condition above implies that for large enough  $N$ ,  $\frac{C_{N+2}}{A_{N+2}} \ll \frac{1}{A_N}$  and we can ignore the contribution from the first term in the square bracket, resulting in:  $\bar{\rho}_{N+1}(t) \approx \frac{-i}{\hbar \lambda_{N+1} A_N} V^\times \bar{\rho}_N(t)$ . Using this approximation, we can terminate the hierarchy at level  $N$  and formulate the *terminator* equation as:

$$\frac{\partial}{\partial t} \bar{\rho}_N(t) = - \left[ \frac{i}{\hbar} \left( H_0^\times - \frac{B_N}{A_N} V^\times \right) + \lambda_N \right] \bar{\rho}_N(t) - \frac{1}{\hbar^2} \frac{C_{N+1}}{\lambda_{N+1} A_{N+1} A_N} V^\times V^\times \bar{\rho}_N(t) - \frac{i}{\hbar} \frac{1}{A_{N-1}} V^\times \bar{\rho}_{N-1}(t) \quad (\text{B2})$$

### Appendix C: Explicit forms of the DHEOMs

In this Appendix we explicitly write DHEOM for quantum systems driven by the three diffusive Markov processes that permit such descriptions. We have confirmed that these DHEOM descriptions of dynamical evolution agree with averaged Monte-Carlo simulations of the same noise processes.

#### 1. Ornstein-Uhlenbeck process

For the case where  $\Omega(t)$  is an Ornstein-Uhlenbeck process, Kubo and Tanimura [5, 9] have demonstrated how to construct hierarchical equations of motion for the reduced density matrix. In fact, the generalization described in this paper was originally motivated by this work. In this section we restate the Kubo-Tanimura DHEOM for the OU process in the present notation.

The joint distribution is first expanded in terms of the eigenfunctions of the Markov generator for the Ornstein-Uhlenbeck process:

$$\hat{\rho}(t, \Omega) \equiv \hat{\rho}_\Omega(t) P(\Omega, t) = \sum_{n=0}^{\infty} \bar{\rho}_n(t) \phi_n(\Omega) \quad (\text{C1})$$

$\phi_n(\Omega) = P_0^{\text{OU}}(\Omega) \overleftarrow{\phi}_n(\Omega)$  are eigenfunctions of the forward generator for the OU process. These quantities are defined explicitly in Table I. Since  $\phi_n(\Omega)$  are proportional to Hermite polynomials a three term recurrence relation they follow is:

$$\Omega \phi_n(\Omega) = \sqrt{\frac{\sigma^2}{2\gamma}} \phi_{n-1}(\Omega) + \mu \phi_n(\Omega) + (n+1) \sqrt{\frac{\sigma^2}{2\gamma}} \phi_{n+1}(\Omega) \quad (\text{C2})$$

where Table I should be referred to for the interpretation of the parameters in this expression. Given this recurrence relation, the resulting DHEOM is:

$$\begin{aligned} \frac{\partial}{\partial t} \bar{\rho}_0(t) &= -\frac{i}{\hbar} (H_0^\times + \mu V^\times) \bar{\rho}_0(t) - \frac{i}{\hbar} \sqrt{\frac{\sigma^2}{2\gamma}} V^\times \bar{\rho}_1(t), \\ \frac{\partial}{\partial t} \bar{\rho}_n(t) &= - \left[ \frac{i}{\hbar} (H_0^\times + \mu V^\times) + n\gamma \right] \bar{\rho}_n(t) - \frac{i}{\hbar} \sqrt{\frac{\sigma^2}{2\gamma}} V^\times \bar{\rho}_{n+1}(t) - \frac{i}{\hbar} n \sqrt{\frac{\sigma^2}{2\gamma}} V^\times \bar{\rho}_{n-1}(t), \quad n > 0 \end{aligned} \quad (\text{C3})$$

The terminator equation is explicitly,

$$\frac{\partial}{\partial t} \bar{\rho}_N(t) = - \left[ \frac{i}{\hbar} (H_0^\times + \mu V^\times) + N\gamma \right] \bar{\rho}_N(t) - \frac{1}{\hbar^2 \gamma} \left( \frac{\sigma^2}{2\gamma} \right) V^\times V^\times \bar{\rho}_N(t) - \frac{i}{\hbar} N \sqrt{\frac{\sigma^2}{2\gamma}} V^\times \bar{\rho}_{N-1}(t) \quad (\text{C4})$$

#### 2. Square-root process

We will expand the joint distribution for the quantum and stochastic variables as

$$\hat{\rho}(t, \Omega) = \sum_{n=0}^{\infty} \bar{\rho}_n(t) \chi_n(\Omega) \quad (\text{C5})$$



where  $\chi_n(\Omega) = P_0^{\text{SR}}(\Omega) \overleftarrow{\chi}_n(\Omega)$  is the expression for the eigenfunctions of the forward generator for the square-root process in terms of the quantities in Table I. The three-term recurrence formula satisfied by the basis functions  $\chi_n(\Omega)$  (which follows from a three-term recurrence relation followed by the Laguerre polynomials) that we require is:

$$\begin{aligned} \Omega \chi_n(\Omega) = & -\frac{c_1}{2\gamma} \frac{(\alpha + n)}{n} \chi_{n-1}(\Omega) \\ & + \left[ \frac{c_1}{2\gamma} (\alpha + 2n + 1) - \frac{c_0}{c_1} \right] \chi_n(\Omega) \\ & - \frac{c_1}{2\gamma} (n + 1)^2 \chi_{n+1}(\Omega) \end{aligned} \quad (\text{C6})$$

where  $\alpha \equiv \frac{2\gamma}{c_1} \left( \mu + \frac{c_0}{c_1} \right) - 1$ , and  $\gamma, \mu, c_0$ , and  $c_1$  are the defining parameters for the process (see Table I). Using this recurrence relation, the resulting hierarchy of coupled equations of motion are:

$$\begin{aligned} \frac{\partial}{\partial t} \bar{\rho}_0(t) = & -\frac{i}{\hbar} \left( H_0^\times + \left[ \frac{c_1}{2\gamma} (\alpha + 1) - \frac{c_0}{c_1} \right] V^\times \right) \bar{\rho}_0(t) + \frac{i}{\hbar} \frac{c_1}{2\gamma} (\alpha + 1) V^\times \bar{\rho}_1(t), \\ \frac{\partial}{\partial t} \bar{\rho}_n(t) = & -\left[ \frac{i}{\hbar} \left( H_0^\times + \left[ \frac{c_1}{2\gamma} (\alpha + 2n + 1) - \frac{c_0}{c_1} \right] V^\times \right) + n\gamma \right] \bar{\rho}_n(t) \\ & + \frac{i}{\hbar} \frac{c_1}{2\gamma} \frac{(\alpha + n + 1)}{n + 1} V^\times \bar{\rho}_{n+1}(t) + \frac{i}{\hbar} \frac{c_1}{2\gamma} n^2 V^\times \bar{\rho}_{n-1}(t), \quad n > 0 \end{aligned} \quad (\text{C7})$$

The terminator equation is explicitly,

$$\begin{aligned} \frac{\partial}{\partial t} \bar{\rho}_N(t) = & -\left[ \frac{i}{\hbar} \left( H_0^\times + \left[ \frac{c_1}{2\gamma} (\alpha + 2N + 1) - \frac{c_0}{c_1} \right] V^\times \right) + N\gamma \right] \bar{\rho}_N(t) \\ & - \frac{1}{\hbar^2 \gamma} \left( \frac{c_1}{2\gamma} \right)^2 (\alpha + N + 1) V^\times V^\times \bar{\rho}_N(t) + \frac{i}{\hbar} \frac{c_1}{2\gamma} N^2 V^\times \bar{\rho}_{N-1}(t) \end{aligned} \quad (\text{C8})$$

Note that this choice of recurrence relation leads to the coefficient of  $V^\times$  scaling as  $\rightarrow n/\gamma$  for large  $n$ . At the same time, the eigenvalue of the generator scales as  $\rightarrow \gamma n$ . This means that the first condition for the existence of a truncation strategy, that  $|\lambda_N| \gg \|H_0^\times - \frac{B_N}{A_N} V^\times\|_2$  can only hold true for  $\gamma > 1$ . Therefore we are only guaranteed that a valid truncation exists when  $\gamma > 1$  for the square-root process. It is possible that this condition can be lifted by using an alternate recurrence relation for the generalized Laguerre polynomials but we have not been able to find such a recurrence relation at this time, and therefore are limited to simulating square-root processes with  $\gamma > 1$  using the above DHEOM.

### 3. Jacobi process

If the stochastic driving of the quantum system is by a Jacobi process, we expand the joint distribution in terms of the eigenfunctions of its generator

$$\hat{\rho}(t, \Omega) = \sum_{n=0}^{\infty} \bar{\rho}_n(t) \zeta_n(\Omega) \quad (\text{C9})$$

where  $\zeta_n(\Omega) = P_0^{\text{J}}(\Omega) \overleftarrow{\zeta}_n(\Omega)$  in terms of the quantities defined in Table I. The three-term recurrence formula we will use, which follows from a three-term recurrence formula of Jacobi polynomials, is:

$$\begin{aligned} \Omega \zeta_n(\Omega) = & \frac{\Delta\omega}{2} \left( \frac{(\alpha + n)(\beta + n)}{n\eta_n(\eta_n + 1)} \right) \zeta_{n-1}(\Omega) \\ & + \left[ \omega_2 - \frac{\Delta\omega}{2} \left( \frac{\alpha^2 - \beta^2}{\eta_n(\eta_n + 2)} + 1 \right) \right] \zeta_n(\Omega) \\ & + \frac{\Delta\omega}{2} \left( \frac{(n + 1)^2(\eta_n - n + 1)}{(\eta_n + 1)(\eta_n + 2)} \right) \zeta_{n+1}(\Omega) \end{aligned} \quad (\text{C10})$$

where  $\alpha$  and  $\beta$  are as defined in Table I, and  $\Delta\omega \equiv \omega_2 - \omega_1$ , and  $\eta_n \equiv \alpha + \beta + 2n$ . Using this recurrence relation, we derive the following DHEOM:

$$\begin{aligned} \frac{\partial}{\partial t} \bar{\rho}_0(t) &= -\frac{i}{\hbar} \left( H_0^\times + \left[ \omega_2 - \frac{\Delta\omega}{2} \left( \frac{\alpha^2 - \beta^2}{\eta_0(\eta_0 + 2)} + 1 \right) \right] V^\times \right) \bar{\rho}_0(t) - \frac{i}{\hbar} \frac{\Delta\omega}{2} \left( \frac{(\alpha + 1)(\beta + 1)}{\eta_1(\eta_1 + 1)} \right) V^\times \bar{\rho}_1(t) \\ \frac{\partial}{\partial t} \bar{\rho}_n(t) &= -\left[ \frac{i}{\hbar} \left( H_0^\times + \left[ \omega_2 - \frac{\Delta\omega}{2} \left( \frac{\alpha^2 - \beta^2}{\eta_n(\eta_n + 2)} + 1 \right) \right] V^\times \right) + (n\gamma + \frac{1}{2}cn(n-1)) \right] \bar{\rho}_n(t) \\ &\quad - \frac{i}{\hbar} \Delta\omega \left( \frac{(\alpha + n + 1)(\beta + n + 1)}{(n + 1)\eta_{n+1}(\eta_{n+1} + 1)} \right) V^\times \bar{\rho}_{n+1}(t) \\ &\quad - \frac{i}{\hbar} \Delta\omega \left( \frac{n^2(\eta_{n-1} - n + 2)}{(\eta_{n-1} + 1)(\eta_{n-1} + 2)} \right) V^\times \bar{\rho}_{n-1}(t), \quad n > 0 \end{aligned} \quad (C11)$$

The terminator equation for this DHEOM is explicitly,

$$\begin{aligned} \frac{\partial}{\partial t} \bar{\rho}_N(t) &= -\left[ \frac{i}{\hbar} \left( H_0^\times + \left[ \omega_2 - \frac{\Delta\omega}{2} \left( \frac{\alpha^2 - \beta^2}{\eta_N(\eta_N + 2)} + 1 \right) \right] V^\times \right) + (N\gamma + \frac{1}{2}cN(N-1)) \right] \bar{\rho}_N(t) \\ &\quad - \frac{1}{\hbar^2(\gamma + \frac{c}{2}N)} \Delta\omega^2 \left( \frac{(\alpha + N + 1)(\beta + N + 1)(\eta_N - N + 1)}{\eta_{N+1}(\eta_{N+1} + 1)(\eta_{N+1} + 2)} \right) V^\times V^\times \bar{\rho}_N(t) \\ &\quad - \frac{i}{\hbar} \Delta\omega \left( \frac{N^2(\eta_{N-1} - N + 2)}{(\eta_{N-1} + 1)(\eta_{N-1} + 2)} \right) V^\times \bar{\rho}_{N-1}(t) \end{aligned} \quad (C12)$$

It is interesting to note that the growth of the  $\lambda_n$  with  $n$  for the Jacobi process is quadratic (as opposed to linear for the OU and square-root processes). This implies that the termination conditions for the DHEOM are met much more quickly for the Jacobi process, and hence it is more efficient to simulate than the other processes.

#### Appendix D: DHEOM for the integrated map

In the main text, we derived a general DHEOM for the density matrix describing a quantum system driven by a diffusive Markov process. In solving for the dynamics of a quantum systems, it is often useful to work at the level of an integrated (completely positive) map, or propagator, rather than a density matrix. The two are related simply as  $\rho(t) = \mathcal{E}(t)\rho^0$ , where  $\mathcal{E}(t)$  is the integrated map that propagates any initial state  $\rho^0$  to  $\rho(t)$ . If one chooses to vectorize the  $n \times n$  density matrix into an  $n^2 \times 1$  vector by stacking the columns:  $\vec{\rho} = \text{vec}(\hat{\rho})$ , then  $\mathcal{E}(t)$  is a matrix of size  $n^2 \times n^2$ . For unitary evolution, where  $\rho(t) = U(t)\rho_0 U^\dagger(t)$ , we get a matrix representation  $\mathcal{E}(t) = U^*(t) \otimes U(t)$  from the identity  $\text{vec}(ABC) = (C^T \otimes A)\text{vec}(B)$  for any matrices  $A, B, C$  [22]. Here  $*$  denotes complex conjugation and  $^T$  denotes transposition. The Schrödinger (von Neumann) equation describing evolution by a Hamiltonian  $\hat{H}(t)$  translates to

$$\frac{\partial}{\partial t} \mathcal{E}(t) = -\frac{i}{\hbar} \left[ \hat{1} \otimes \hat{H}(t) - \hat{H}^T(t) \otimes \hat{1} \right] \mathcal{E}(t) \equiv -\frac{i}{\hbar} \mathcal{H}(t) \mathcal{E}(t), \quad (D1)$$

with initial condition  $\mathcal{E}(0) = \mathbf{I}$ . In this appendix, we develop the generalized DHEOM that describes the reduced equations of motion for these propagators.

Similar to the density matrix derivation, we begin by defining  $\mathcal{E}(t, \Omega) \equiv \mathcal{E}_\Omega(t)P(\Omega, t)$  where  $\mathcal{E}_\Omega$  is the conditioned propagator  $\mathcal{E}_\Omega(t) \equiv \mathcal{E}(t|\Omega(t))$ . This quantity satisfies a propagator version of the stochastic Liouville equation:

$$\frac{\partial \mathcal{E}(t, \Omega)}{\partial t} = \left[ -\frac{i}{\hbar} \mathcal{H}_0 - \frac{i}{\hbar} \Omega(t) \mathcal{V} \right] \mathcal{E}(t, \Omega) + \Gamma_\Omega \mathcal{E}(t, \Omega), \quad (D2)$$

where we have split the Hamiltonian into a time-independent component and a Markov fluctuating component:  $\mathcal{H} = \mathcal{H}_0 + \Omega(t)\mathcal{V}$ . As with the density matrix, the averaged propagator is given by the marginal:

$$\mathcal{E}(t) = \int_{\mathcal{D}} d\Omega \mathcal{E}(t, \Omega). \quad (D3)$$

When  $\Omega(t)$  is a diffusive process with a generator possessing polynomial eigenfunctions, we expand  $P(\Omega, t)$  in generator eigenfunctions  $f_n(\Omega)$  as

$$\mathcal{E}(t, \Omega) \equiv \mathcal{E}_\Omega(t)P(\Omega, t) = \sum_{n=0}^{\infty} \bar{\mathcal{E}}_n(t) f_n(\Omega). \quad (D4)$$

Then utilizing the three-term recurrence relations that the orthogonal polynomial eigenfunctions follow, we arrive at the following classical hierarchical equations of motion for the propagator:

$$\begin{aligned}\frac{\partial}{\partial t}\bar{\mathcal{E}}_0(t) &= -\left[\frac{i}{\hbar}\left(\mathcal{H}_0 - \frac{B_0}{A_0}\mathcal{V}\right) + \lambda_0\right]\bar{\mathcal{E}}_0(t) - \frac{i}{\hbar}\frac{C_1}{A_1}\mathcal{V}\bar{\mathcal{E}}_1(t), \\ \frac{\partial}{\partial t}\bar{\mathcal{E}}_n(t) &= -\left[\frac{i}{\hbar}\left(\mathcal{H}_0 - \frac{B_n}{A_n}\mathcal{V}\right) + \lambda_n\right]\bar{\mathcal{E}}_n(t) - \frac{i}{\hbar}\frac{C_{n+1}}{A_{n+1}}\mathcal{V}\bar{\mathcal{E}}_{n+1}(t) - \frac{i}{\hbar}\frac{1}{A_{n-1}}\mathcal{V}\bar{\mathcal{E}}_{n-1}(t), \quad n > 0,\end{aligned}\quad (\text{D5})$$

where  $\lambda_n, A_n, B_n$  and  $C_n$  are eigenvalue and eigenfunction recurrence relation coefficients defined identically as in the main text. The terminator equation for this DHEOM is explicitly:

$$\frac{\partial}{\partial t}\bar{\mathcal{E}}_N(t) = -\left[\frac{i}{\hbar}\left(\mathcal{H}_0 - \frac{B_N}{A_N}\mathcal{V}\right) + \lambda_N\right]\bar{\mathcal{E}}_N(t) - \frac{1}{\hbar^2}\frac{C_{N+1}}{\lambda_{N+1}A_{N+1}A_N}\mathcal{V}\mathcal{V}\bar{\mathcal{E}}_N(t) - \frac{i}{\hbar}\frac{1}{A_{N-1}}\mathcal{V}\bar{\mathcal{E}}_{N-1}(t). \quad (\text{D6})$$


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